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Mr. Everett A. Wick
High-Level Waste Licensing Management Branch
Division of Waste Management
Office of Nuclear Material Safety & Safeguards
Mail Stop 623-SS
U. S. Nuclear Regulatory Commission
Washington, D. C. 20555

Dear Mr. Wick:

This is the monthly management report for the month of October for the programs entitled, "Review of DOE Waste Package Program," FIN A-3164 and "Waste Package Verification Tests," FIN A-3167. Included are the monthly highlight letters for the aforementioned programs. The breakdown of costs by task is given on the attached computer summary sheets. Projections of costs by task are given for FY 1985.

If there are any questions regarding format, distribution, or budget reporting, please contact Mr. A. J. Weiss, Administrative Technical Assistant, FTS 666-4473.

Sincerely yours,

Walter Y. Kato
Deputy Chairman

WYK/jw

Enclosures

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REVIEW OF DOE WASTE PACKAGE PROGRAM
(FIN A-3164)

P. Soo

Monthly Letter Report, October, 1985
Published: November, 1985

Nuclear Waste Management Division
Department of Nuclear Energy
BROOKHAVEN NATIONAL LABORATORY
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Task 1 - Review of DOE Waste Package Program

The formal review of the DOE waste package program, carried out on a continuing basis over the last six years, has been terminated. Future work in this effort will be in the form of general technical assistance to the NRC.

Review of "Thermal Considerations for Emplaced Waste Packages" (T. Sullivan)

The subject NRC report addresses the problem of estimating temperature distributions around emplaced waste packages. Calculations were carried out by NRC staff based on specific simplifying assumptions and their relevance to waste package performance was discussed. A review of the study was carried out at the request of NRC and the findings were sent to the program manager under separate cover.

Attendance at the ACRS Meeting on October 24-25, 1985 (T. Sullivan, P. Soo)

Staff from BNL attended the subject meeting in which NRC personnel presented outlines of programs concerned with high-level waste management.

REVIEW OF WASTE PACKAGE VERIFICATION TESTS
(FIN A-3167)

P. Soo

Monthly Letter Report, October, 1985
Published: November, 1985

Nuclear Waste Management Division
Department of Nuclear Energy
BROOKHAVEN NATIONAL LABORATORY
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Task 1 - Review of Waste Package Verification Tests (T. Abraham, H. Jain, T. Sullivan, C. Anderson, and P. Soo)

The final version of the semi-annual report for April-September 1985, has been completed. A camera-ready copy has been sent for patent clearance after which it will be forwarded to NRC for printing.

This will be the last semi-annual report (Volume 7) in the series since the program is being terminated. Work in the immediate future will involve completion of the Task 3 work on the evaluation of stainless steel stress corrosion cracking, and work in the General Technical Assistance area (Task 4).

Task 2 - Modeling/Code Evaluation (T. Sullivan)

Evaluation of the Ingrowth of Radionuclides After the Containment Period and the Impact on Meeting Controlled Release Limits

Ingrowth is the process by which the concentraton of a radionuclide may increase with time due to the radioactive decay of other elements. A memorandum covering this subject has been completed and is attached as an Appendix. The conclusions of this study are, that for spent fuel, ingrowth of radionuclides will not present a problem in terms of the controlled release criteria. The reason for this is that none of the radionuclides that exhibit ingrowth has an inventory that exceeds one part in 10^5 of the total 1,000-year inventory. Thus, their release is restricted to less than one part in 10^8 of the 1,000-year total radionuclide inventory and does not change due to ingrowth.

Response to Comments on the Generic Technical Position (GTP) on Waste Package Reliability Analysis

The Waste Management Branch of NRC's Division of Radiation Programs and Earth Sciences reviewed the subject BNL document. Their review was extremely thorough and the comments helped to clarify the meaning of the GTP. The reviewer(s) should be commended for their excellent work. The response to their comments was sent to NRC under separate cover for inclusion in a finalized report.

Task 3 - Stress Corrosion Cracking Studies on Container Materials for a Tuff Repository (T. Abraham, C. Anderson, P. Soo)

The objective of this test program is to evaluate the stress corrosion cracking susceptibility of the NNWSI candidate container alloys, viz. Types 304L, 316L, and 321 stainless steel, and Incoloy 825, in a simulated tuff repository environment. The V-notched, C-ring test method has been adopted in which each specimen (used in triplicate for each test condition) is stressed to 90% of the elastic limit calculated for the unnotched condition. Stressed specimens are exposed at ≈100°C to the liquid as well as the steam

phase over synthetic J-13 groundwater and ten-times-concentrated J-13 groundwater; both in contact with crushed Topopah Spring tuff. The use of concentrated water simulates the situation when salts, precipitated after initial evaporation of groundwater, are redissolved in cooler water subsequently percolating towards the repository horizon.

The final test solutions from the 12-month C-ring tests in J-13 and ten-times concentrated J-13 solutions have been analyzed and are shown in Table 1. Results for both tests show that dilution by a factor of two reduces the concentration approximately by a factor of two (compare Sample No. 19 with No. 18 and Sample No. 21 with 20). As in the cases of three-month and six-month tests, no significant precipitation appears to occur during the cooling of undiluted solutions. To confirm whether any precipitation occurs in the presence of tuff rocks at room temperature, 150 mL of ten-times concentrated J-13 solution was cooled to room temperature and it was maintained in contact with the tuff for three days. The solution was filtered and analyzed. The analysis is shown in Table 1 (Sample No. 22). Comparing it with the undiluted and filtered solution (Sample No. 21), it is seen that the concentrations of various species in both solutions are the same, within experimental error, except for SiO_2 which decreased by 40% from the original concentration. Therefore, a part of SiO_2 in the solution may have been removed as particulate matter.

Table 2 gives the compositions of the solutions after three-month, six-month and one-year tests. Composition of reference J-13 groundwater is included for the purpose of comparison. The concentration of the species in the solution at the conclusion of all three tests increased many-fold compared to the starting solution. However, as the tests progress from three-month to one-year, there is no trend in the variation of concentrations of the aqueous species except for SO_4^{2-} and Ca^{2+} ions, both of which decrease with time. This could be attributed to the precipitation of $CaSO_4$ which may have adhered to the surface of the specimens or the tuff. The salt film on the surface of a Type 304L specimen in a six-month test in J-13 water underwent an energy dispersive X-ray analysis (EDAX) to determine its composition (see Monthly Letter Report, June 1985). Qualitatively, silicon and aluminum were identified as the major elements with calcium, chloride and sulfur present in smaller concentrations. This supports the proposed precipitation of $CaSO_4$ from the solution. Table 2 also shows that the pH increases from 8.4 in the three-month tests to about 9.3 in the one-year tests. An earlier investigation by one of the authors (T. Abraham)[a] which showed that the stability region of $CaSO_4$ increases with increasing pH as seen in this investigation.

Response to NRC Comments on the July Monthly Letter Report

NRC requested clarification of some of the work in this Task reported in the July 1985 Monthly Letter Report. In particular, BNL was asked to describe the machining of the V notches in the C-ring samples with respect to the use or non-use of lubricants, the depth of the cold worked layer, and the possibility of introducing microcracks during the machining processes.

[a]Abraham, Thomas, "A Thermodynamic Study of the Aqueous System, Ca-S-H_2O," Doctoral Thesis, Columbia University, New York, 1981.

Table 1. Chemical composition of test solutions at the end of the one-year tests (μg/mL)

	Reference J-13 Groundwater	Synthetic J-13 Water		10-Times Conc. J-13-Water		
		Sample 19	Sample 18	Sample 21	Sample 20	Sample 22
Na^+	45	510	280	908	472	897
K^+	4.9	106	64	139	80.5	83
Ca^{2+}	14	104	48.4	129	54	134
Sn^{2+}		1.0	0.5	1.2	0.5	1.7
F^-	2.2	6.3	2.0	21.1	4.6	9.3
Cl^-	7.5	161	80	260	108	255
NO_3^-	5.6	482	247	672	339	660
SO_4^{2-}	22	588	298	976	489	966
SiO_2	61	458	276	406	216	240
pH at room temperature	8.5	9.3 8.5 (at 101°C)		9.3 8.6 (at 100°C)		

Sample 18 diluted (2x) and filtered.

Sample 19 undiluted and filtered.

Sample 20 diluted (2x) and filtered.

Sample 21 undiluted and filtered.

Sample 22 solution cooled and contacted with tuff, and filtered.

Table 2. Chemical composition of test solutions at the end of corrosion tests (μg/mL) (undiluted and filtered solution).

	Reference J-13 Groundwater	Synthetic J-13 Water			10-Times Conc. J-13-Water		
		3-month Test	6-month Test	1-year Test	3-month Test	6-month Test	1-year Test
Na^+	45	N.D.*	464	510	867	738	908
K^+	4.9	238	244	106	244	214	139
Ca^{2+}	14	308	161	104	301	164	129
Sn^{2+}		3.4	0.4	1.0	4.4	0.5	1.2
F^-	2.2	12.1	4	6.31	14	5	21.1
Cl^-	7.5	130	236	161	330	211	260
NO_3^-	5.6	460	750	482	------	522	672
SO_4^{2-}	22	820	552	588	1300	1260	976
SiO_2	61	414	451	458	408	488	406
pH at room temperature	8.5	8.4	9.0	9.3	8.4	8.9	9.3

*N.D. = not determined

The notches were produced by milling and regular machine oil was used for lubrication. Specimens were carefully degreased in acetone and alcohol prior to testing. From metallographic examination after testing the cold worked layers were approximately five micrometers in thickness. It is highly unlikely that the microcracks, seen in the Type 316L stainless steel exposed to the air/steam environment above 10X concentrated J-13 water and tuff for six months, were caused by the machining of the notch since austenitic stainless steels are extremely ductile and difficult to crack by small-scale milling procedures. In addition, it should be noted that the microcracks are parallel to the longitudinal direction of the notch and, therefore, should not be induced by surface smearing of material by machining. This is one reason why we do not believe that a detailed examination of as-machined (control) specimens, which was suggested by NRC, is worthwhile. Also, it should be recognized that the several specimens which we have currently found to contain microcracks were

exposed to the steam/air environment and not boiling solutions. This gives a clear indication that they are a result of the test environment rather than the V-notch-machining process.

Finally, it should be recognized that the current C-ring test methodology, although a standard ASTM test to detect the susceptibility of a stainless steel to stress-corrosion cracking, is quite realistic from a waste management standpoint since a container could be inadvertently scratched during handling or emplacement leaving a notch with a deformed surface layer. A C-ring sample, similar to those used in the BNL study, could be used to simulate this condition. It would be inappropriate to machine a V-notch and anneal the specimen to remove the cold-worked layer.

APPENDIX

BROOKHAVEN NATIONAL LABORATORY

MEMORANDUM

DATE: November 5, 1985

TO: Files - NWM-MF-#34

FROM: T. Sullivan T.S.

SUBJECT: Ingrowth of Radionuclides in High-Level Spent Fuel Waste and the Effect on Controlled Release Criteria

1.0 Introduction

The NRC Rule (10 CFR Part 60) for disposal of high level radioactive wastes states that the maximum permissable release rates for any radionuclide shall not exceed one part in 100,000 per year of the inventory of that radionuclide calculated to be present 1000 years after permanent closure. This requirement does not apply to any radionuclide which is released at a rate of less than 0.1% of the calculated total release rate limit which is defined as one part in 100,000 per year of the inventory of radioactive waste emplaced in the underground facility that remains after 1000 years of radioactive decay. This requirement reduces to having a release of less than one part in 10^8 of the 1000 year inventory. Under special circumstances the Commission may approve of different maximum release rates.

The release rate limits are based on the 1000 year inventory and are not oriented to consider the situation that arises when ingrowth occurs. Ingrowth is the increase in inventory over time that occurs when the production of a radionuclide due to radioactive decay of other nuclides exceeds the destruction of the nuclide under consideration due to its own decay. For radionuclides that exhibit ingrowth after 1000 years, the increased inventory would make the controlled release criterion of one part in 100,000 of the 1000 year inventory more difficult to meet. If the nuclides exhibiting ingrowth fall under the controlled release criterion of 1 part in 10^8 of the total inventory at 1000 years, ingrowth will not pose a special problem. This memorandum lists the important decay chains and, using a typical spent fuel inventory, explores the potential problems in meeting controlled release criteria for nuclides that demonstrate ingrowth.

2.0 Radionuclide Inventories and Decay Chains

The first piece of information required to determine if ingrowth of radionuclides will present a problem to controlled release is the typical inventory expected in a repository. A spent fuel inventory is presented in Table 1 for several of the actinides at times of 0, 200, and 1000 years after disposal. The inventories of the fission products are not presented because ingrowth of these nuclides is not a problem on the time scale of thousands of years.

Table 1 Radionuclide inventories from 72,000 MTHM of PWR spent fuel up to 1000 years.

	Inventory (Curies)		
Radionuclide	0 yr	200 yr	1000 yr
Ra-226	2.6×10^{-2}	8.7×10^{0}	2.3×10^{2}
Th-230	9.5×10^{0}	2.1×10^{2}	1.2×10^{3}
U -233	1.9×10^{0}	2.8×10^{1}	2.3×10^{2}
U -234	8.6×10^{4}	1.3×10^{5}	1.5×10^{5}
U -238	2.3×10^{4}	2.3×10^{4}	2.3×10^{4}
Np-237	2.2×10^{4}	3.8×10^{4}	7.1×10^{4}
Pu-238	1.6×10^{8}	3.4×10^{7}	7.1×10^{4}
Pu-239	2.3×10^{7}	2.2×10^{7}	2.2×10^{7}
Pu-240	3.8×10^{7}	3.7×10^{7}	3.4×10^{7}
Pu-241	5.5×10^{9}	5.2×10^{5}	9.2×10^{3}
Pu-242	1.2×10^{5}	1.2×10^{5}	1.2×10^{5}
Am-241	1.2×10^{8}	2.3×10^{8}	6.3×10^{7}
Am-243	1.2×10^{6}	1.2×10^{6}	1.1×10^{6}
Cm-244	9.1×10^{7}	4.3×10^{4}	≈0
Cm-245	10,000	9.8×10^{3}	9.2×10^{3}

The data for the spent fuel inventory were obtained from the Richton Dome Draft Environmental Assessment, Table 6.27. These data were originally obtained from use of the ORIGEN computer code with the assumption that there are 72,000 MTHM of PWR spent fuel. It should be noted that these data appeared in all seven salt site draft Environmental Assessments and is expected to be a typical repository inventory. Currently, there is not a standard radionuclide inventory for any of the waste forms or are there standard half-lives for the radionuclides. There were differences of up to five percent between the inventories and half-lives used in the salt, basalt, and tuff environmental assessments for performance analysis. Standardization of the inventories similar to the ANS decay heat curves would clear up these small discrepancies and provide a uniform starting point for controlled release calculations.

From Table 1, it is seen that Ra-226 and Th-230 inventories increase by a few orders of magnitude over the first 1000 years. The inventories of U-233,

U-234, and Np-237 also increase during this time period. Also, the inventory of Am-241 increases over the first few hundred years before decreasing.

To understand this behavior requires an inspection of the radioactive decay chains and half-lives presented in Tables 2 through 5 and some fundamental information on ingrowth.

Table 2 Decay chain beginning with Pu-242 or Pu-238.

Nuclide	Half-Life	Decay Mode
Pu-238	87.8 y	α
U -234	2.44×10^5 y	α
Th-230	7.7×10^4 y	α
Ra-226	1600 y	α
Rn-222	3.8 d	α
Po-218	3 m	α
Pb-214	27 m	β^-
Bi-214	20 m	β^-
Po-214	167 μs	α
Pb-210	22.3 y	β^-
Bi-210	5 d	β^-
Po-210	138 d	α
Pb-206	stable	α
An alternate route to enter this chain at U-234 is		
Pu-242	3.9×10^5 y	α
U -238	4.5×10^9 y	α
Th-234	24.1 d	β^-
Pa-234	6.7 h	β^-
U -234	2.44×10^5 y	α

2.1 Discussion of Ingrowth of Radionuclides

Ingrowth is likely to occur when a member of a decay chain has a long half life as compared to the preceding members of the chain or if the initial inventory of the radionuclide is small.

As an example, consider the decay chain in Table 2. Pu-238 with an 87.8 yr half-life decays into U-234 with a 2.4×10^5 yr half-life and thereby causes the ingrowth of U-234. Eventually, the U-234 decays into Th-230. Although the half-life of Th-230 is much less than U-234, the initial inventory of Th-230 is essentially zero and thus the production of Th-230 exceeds the destruction and the inventory increases for some time.

It should be recognized that upon emplacement of spent fuel in the repository, of the nuclides listed in Tables 2-5 only those with an atomic number greater than 91 (U, Np, Pu, Am, and Cm) will exist in any significant amount. This is important because for radionuclides with an atomic number of 91 or less it implies that their maximum activity will never exceed the smallest activity of the radionuclide that precedes them in the decay chain. That is, 1 curie of U-234 becomes at most 1 curie of Th-230.

Table 3 Decay chain beginning with Am-243.

Nuclide	Half-Life	Decay Mode
Am-243	7370 y	α
Np-239	2.3 d	β^-
Pu-239	2.4×10^4 y	α
U -235	7×10^8 y	α
Th-231	25 h	β^-
Pa-231	3.2×10^4 y	α
Ac-227	21.8 y	β^-
Th-227	18.7 d	α
Ra-223	11.4 d	α
Rn-219	3.9 s	α
Po-215	0.002 s	α
Pb-211	36.1 m	β^-
Bi-211	2.1 m	α
Tl-207	4.8 m	β^-
Pb-207	stable	

Another important point to realize is that activity is defined as the product of the decay constant and the number of atoms of the radionuclide. Thus, radionuclides with a large decay constant may have a large activity without having a large mass inventory. For example, from Table 1 the curie inventory of Pu-238 is initially 1.6×10^8 curies and the decay constant (ln 2 divided by the half life) is 7.9×10^{-3} yr^{-1}. Eventually, the Pu-238 decays into U-234 which has a decay constant of 2.8×10^{-6} yr^{-1}. If all of the Pu-238 were transformed to U-234 instantly, the 1.6×10^8 curies of Pu-238 would become only 5.75×10^4 curies of U-234. This is less than the initial curie inventory of U-234. Thus, the initial mass inventory of U-234 exceeds that of Pu-238 even though the initial curie inventory of Pu-238 is four orders of magnitude greater than for U-234.

2.2 Discussion of Radioactive Decay Chains

Examination of the decay chains will highlight the sources that lead to ingrowth and with the use of the curie inventories in Table 1 will allow quantification of the magnitude of ingrowth.

Table 2 presents a decay chain that begins with either Pu-242 or Pu-238 and includes U-238, U-234, Th-230, and Ra-226. From this chain and the half lives of the radionuclides in the chain, it is clear that the buildup of U-234 is primarily due to decay of Pu-238 because the half-life of U-238 is 4.5×10^9 years as compared to the 87.8 year half-life of Pu-238. Further, the build-up of Th-230 and Ra-226 results from the decay of U-234.

Table 3 presents a decay chain beginning with Am-243 and includes Pu-239 and U-235. Because of the long half-life of U-235, ingrowth of radionuclides is not a problem for any radionuclides beneath U-235 in the chain. Decay of the entire Am-243 and Pu-239 inventory would result in only 800 curies of U-235.

Table 4 presents a decay chain beginning with Cm-244 which includes Pu-240, U-236, and Th-232. Again, the long half-lives of both U-236 and Th-232 prevent ingrowth of these nuclides or those beneath them in the decay chain from becoming a problem. Ingrowth of Pu-240 could occur due to decay of Cm-244, however, for the inventories expected in spent fuel this will not be a problem as evidenced in Table 1. Further, the half-life of Cm-244 is only 17.9 yr. Thus, any ingrowth would occur before 1000 years and Pu-240 buildup will not be a problem.

Table 5 presents a decay chain beginning with Cm-245 and including Pu-241, Am-241, Np-237, U-233 and Th-229. Due to the initial inventories of Cm-245, Pu-241, Am-241 and the long half-life of Np-237 it is clear that there will be a significant ingrowth of Np-237. Radionuclides below Np-237 in the decay chain will be limited by the decay of Np-237 but ingrowth will occur.

In Table 1 the curie inventory of Np-237 grows from an initial value of 2.2 x 10^4 curies to 7.1 x 10^4 curies after 1000 years. Similarly, the U-233 curie inventory grows from 19 to 230 curies in that time span. The inventory of Np-237 will continue to increase after 1000 years due primarily to decay of Am-241.

3.0 Ingrowth of Radionuclides and Release Rate Limits

As stated in the introduction, radionuclide release rate limits are specified in terms of a fractional release of the inventory expected in a repository 1000 years after permanent closure. Thus, a radionuclide whose inventory increases after this time may have more stringent release rate criteria than a radionuclide with a decreasing concentration. To examine this problem requires calculation of the amount of ingrowth that could occur and calculation of the release rate limits.

There are two release rate limits:

(a) The maximum permissable release rate for a radionuclide shall not exceed one part in 100,000 per year of the inventory of that radionuclide calculated to be present 1000 years after permanent closure.

(b) Requirement (a) does not apply to any radionuclide released at a rate of less than 0.1% of the calculated total release rate limit which is defined as 1 part in 100,000 of the total inventory of radioactive waste that remains 1000 years of radioactive decay. Thus, the release rate for these radionuclides is restricted to being less than one part in 10^8 of the 1000 year inventory.

Inspection of requirements (a) and (b) indicate that for any radionuclide subject to (a), ingrowth will require more restrictive release rate limits. However, since the release rate limit of (b) are based on the total inventory, any radionuclide subject to requirement (b) will not have more restrictive release rate limits due to ingrowth. Thus, ingrowth will be a problem only if the inventory of that radionuclide increases after the 1000 year containment period exceeds 1 part in 10^8 of the calculated total 1000 year inventory. For the inventory used in Table 1, the 1000 year inventory is 1.21 x 10^8 curies.

Inspection of the inventories in Table 1 and the decay chains, Tables 2-5, indicate that Np-237, U-234, Th-230, and Ra-226 will present the biggest problems in terms of ingrowth. The following sections will determine the possibility of these radionuclides reaching an inventory of greater than 0.1% of the total 1000 year inventory and thereby determine if ingrowth could cause more restrictive release rate limits.

3.1 Np-237

After 1000 years, the inventory of Np-237 has increased from an initial value of 2.2×10^4 to 7.1×10^4 curies due to decay of Am-241. This is less than 1.21×10^5 curies which is 0.1% of the total 1000 year inventory. Thus, the release rate limit for Np-237 is 1.21 curies per year (1×10^{-8} of the total 1000 year inventory) and ingrowth will not be a problem provided the curie inventory of Np-237 never exceeds 1.21×10^5 curies.

To demonstrate that this is the case, an upper bound for the amount of Np-237 can be calculated by assuming that all of the atoms of the precursors to Np-237 (Am-241, Pu-241, and Cm-245) are converted to Np-237. For example, the 1000 year inventory of Am-241 is 6.3×10^7 curies. To translate this number to curies of Np-237 the following equation is used:

$$\Delta A^{\text{Am-241}}_{\text{Np-237}} = A_{\text{Am-241}} \cdot T_{\text{Am-241}}/T_{\text{Np-237}} \qquad (1)$$

where: ΔA represents activity in curies of Np-237 obtained from Am-241, A represents the 1000 year activity inventory of Am-241, and T represents half-life in years.

Using Equation (1), the 1000 year activity of Am-241 gives an activity of 12,747 curies of Np-237. Using Equation (1) for Cm-245 and Pu-24. and adding all three to the 1000 year activity of Np-237 gives a maximum activity of 8.4×10^4 curies. Thus, ingrowth of Np-237 will not be a problem.

It should be recognized that Equation (1) gives a fairly accurate estimate of the increase in activity in a radionuclide only if the half-life of the radionuclide is much greater than the half-life of its precursor. If this is not the case, Equation (1) may greatly overpredict the increase in activity. In fact, the increase in activity of radionuclides beneath Np-237 in the chain can never exceed the activity of Np-237. Thus, the maximum activity of a daughter of Np-237 can never be greater than its 1000 year inventory plus the maximum Np-237 inventory. Thus, ingrowth is not a problem for all of the nuclides beneath Np-237 as displayed in Table 5, this includes U-233.

3.2 U-234

The initial inventory of U-234 is 8.6×10^4 curies and increases to 1.5×10^5 curies after 1000 years. This exceeds 0.1% of the total 1000 year inventory and thus further ingrowth will be a problem. Examining Table 2 it is seen that there are two paths to reach U-234, by alpha decay of Pu-238 or from the

path beginning with Pu-242 which includes U-238. The build-up of U-234 in the first 1000 years is due almost exclusively to decay of Pu-238. However, after 1000 years almost all of the Pu-238 has decayed. Using Equation (1) it can be shown that further decay of Pu-238 will add at most 25 curies to the inventory of U-234. Thus significant ingrowth of U-234 could occur only from decay of U-238. Assuming that the decay of Th-234, $T_{1/2}$ of 24.1 days, and Pa-234, $T_{1/2}$ of 6.7 hours, can be represented as an instantaneous process, the mass balance equation for U-234 being produced from U-238 becomes:

$$\frac{dN_2}{dt} = \lambda_1 N_1 - \lambda_2 N_2 \qquad (2)$$

where N is the number of atoms,
λ is the decay constant,
subscript 1 refers to U-238, and
subscript 2 refers to U-234.

Recalling that λN is the definition of activity, and from Table 1 at 1000 years, the activity of U-234 is greater than the activity of U-238 it is clear that the number of atoms and therefore activity of U-234 will decrease in time. Thus, ingrowth of U-234 is not a problem.

3.3 Th-230 and Ra-226

Both of these species are daughters of U-234. Therefore, their maximum activity is controlled by the maximum activity of U-234 and could be as large as 1.5×10^5 curies. As discussed earlier, this exceeds 0.1% of the 1000 year inventory and ingrowth would be a problem if the inventories reached this value. To determine if ingrowth does in fact reach this level requires solution of the system of coupled first order differential equations representing the production and destruction of the various nuclides in the decay chain. To include all of the nuclides would be extremely difficult and tedious to perform analytically. Thus some simplifying assumptions will be made. First, because Ra-226 has a shorter half-life than Th-230, the activity of Ra-226 will never exceed that of Th-230. Therefore, if it can be shown that the Th-230 inventory does not exceed 0.1% of the 1000 year inventory it can also be shown for Ra-226. Second, the only members of the chain considered are U-238, U-234, and Th-230. This is equivalent to assuming that the half-lives of Th-234 and Pa-234 are so short that they are in equilibrium with U-238 and that production of U-234 from Pu-238 is negligible (shown to be a good assumption after 1000 years in the previous section). Thus, the inventories at 1000 years are used as the initial values in solution of the problem. The coupled equations are,

$$\frac{dN_1}{dt} = -\lambda_1 N_1 \qquad (3)$$

$$\frac{dN_2}{dt} = \lambda_1 N_1 - \lambda_2 N_2 \qquad (4)$$

$$\frac{dN_3}{dt} = \lambda_2 N_2 - \lambda_3 N_3 \qquad (5)$$

where λ and N have been previously defined,
Subscript 1 refers to U-238,
Subscript 2 refers to U-234, and
Subscript 3 refers to Th-230.

It can be shown that solution of Eqns. (3) through (5) for N_3 written in terms of activites, A, and using the time 1000 years as t=0 gives:

$$A_3(t) = A_3(0)\, e^{-\lambda_3 t}$$

$$+ A_2(0) \frac{\lambda_3}{\lambda_3 - \lambda_2} \left(e^{-\lambda_2 t} - e^{-\lambda_3 t}\right)$$

$$+ A_1(0) \frac{\lambda_2 \lambda_3}{(\lambda_2 - \lambda_1)} \left[\frac{(e^{-\lambda_1 t} - e^{-\lambda_3 t})}{(\lambda_3 - \lambda_1)} - \frac{(e^{-\lambda_2 t} - e^{-\lambda_3 t})}{(\lambda_3 - \lambda_2)} \right] \qquad (6)$$

Here the first term (A_1 in Table 6) represents the activity due to decay of the Th-230 present at t=0, the second and third terms (A_2 and A_3 in Table 6) are the activity of Th-230 produced by U-234, and U-238 respectively. Solution of Equation (6) for various times using the 1000 year inventories as the initial conditions is presented in Table 6.

From Table 6 it is clear that the maximum Th-230 concentration occurs around 2×10^5 years after emplacement and reaches a value of 9.4×10^4 curies. This is below 1.21×10^5 curies which is 0.1% of the 1000 year inventory. Therefore, the more detailed calculation has shown that ingrowth of Th-230 and therefore Ra-226 will not pose a special problem in terms of controlled release rate limits.

The calculations in this memorandum were based on PWR spent fuel removed from the reactor for 10 years. If this is not the initial condition the preceding calculations may need to be repeated as Np-237, Th-230, and Ra-226 all reach approximately 0.08% of the 1000 year inventory. If the waste composition changed due to higher burnup or other factors causing the inventories of any of these radionuclides to exceed 0.1% of the 1000 year inventory, ingrowth will be a problem. If that occurs more thought on the controlled release criteria is necessary.

Table 6 Activity of Th-230 as a function of time. A_T equals $A_3(t)$ in Eqn. 6 and is the sum of the preceding three columns in the Table.

Time After Disposal (yrs)	A_1(curies)	A_2(curies)	A_3(curies)	A_T(curies)
10^3	1.2×10^3	—	—	1.2×10^3
1.1×10^4	1.1×10^3	1.3×10^4	2.8×10^1	1.4×10^4
1.01×10^5	4.9×10^2	7.6×10^4	2.0×10^3	7.8×10^4
2.01×10^5	2.0×10^2	8.8×10^4	5.7×10^3	9.4×10^4
3.01×10^5	8.0×10^1	7.9×10^4	9.4×10^3	8.9×10^4
5.01×10^5	1.3×10^1	5.1×10^4	1.5×10^4	6.6×10^4
1.001×10^6	1.5×10^{-1}	1.2×10^4	2.1×10^4	3.3×10^4